Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

Thermal Analysis for In-tank Ion-Exchange Column Process

Si Y. Lee* and Frank G. Smith, III
Computational Sciences Department
Savannah River National Laboratory
Aiken, SC 2980
**si.lee@srnl.doe.gov ; frank02.smith@srnl.doe.gov*

INTRODUCTION

High Level Waste (HLW) at the Savannah River Site (SRS) is stored in three forms: sludge, saltcake, and supernate. A small column ion-exchange (SCIX) process is being designed to treat dissolved saltcake waste before feeding it to the saltstone facility to be made into grout. The waste is caustic with high concentrations of various sodium salts and lower concentrations of radionuclides. Two cation exchange media being considered are a granular form of crystalline silicotitanate (CST) and a spherical form of resorcinol-formaldehyde (RF) resin. CST is an inorganic material highly selective for cesium that is not elutable.

Through this process, radioactive cesium from the salt solution is absorbed into ion exchange media (either CST or RF) which is packed within a flow-through column. A packed column loaded with radioactive cesium generates significant heat from radiolytic decay. If engineering designs cannot handle this thermal load, hot spots may develop locally which could degrade the performance of the ion-exchange media. Performance degradation with regard to cesium removal has been observed between 50 and 80^{o}C for CST [1] and at 65^{o}C for RF resin [2]. In addition, the waste supernate solution will boil around 130^{o}C. If the columns boiled dry, the sorbent material could plug the column and lead to replacement of the entire column module. Alternatively, for organic resins such as RF there is risk of fire at elevated temperatures.

The objective of the work is to compute temperature distributions across CST- and RF-packed columns immersed in waste supernate under accident scenarios involving loss of salt solution flow through the beds and, in some cases, loss of coolant system flow. For some cases, temperature distributions are determined as a function of time after the initiation of a given accident scenario and in other cases only the final steady-state temperature distributions are calculated. In general, calculations are conducted to ensure conservative and bounding results for the maximum temperatures achievable using the current baseline column design. This information will assist in SCIX design and facility maintenance.

DESCRIPTION OF THE ACTUAL WORK

The baseline column design uses a 28-inch ID cylinder with a 6-inch centerline inner cooling tube and four external cooling jackets prepared from 3.5-inch half-pipe tubes symmetrically distributed around the periphery of the column and oriented along the vertical column height of 15 feet. Coolant water at 25C is supplied at 12.5 gpm to the centerline cooling tube located at its center. Coolant water flow is supplied to each of the outer jackets at 6.25 gpm. For each case the maximum cesium loading was assumed with initial internal column and external air temperatures of 35C.

Equilibrium cesium loadings for each of the CST and RF media were calculated for the projected waste feed streams using the TMIXP code developed at SRNL [3]. Predicted cesium equilibrium loading levels for CST and RF for the most limiting waste type were 257 and 133 Ci Cs-137/L packed bed, respectively [4]. This highly concentrated radioactive source will generate a significant amount of heat in the column, which corresponds to about 5 watts/gallon of volumetric heat source. Under normal operating conditions, process fluid flow through the column can provide adequate heat removal from the column through a coupled conduction and convection heat transfer mechanism. However, in the case of a loss of flow accident, there are concerns about the transient thermal response rates and the maximum steady-state temperatures reached for fully-loaded columns containing each ion exchange media. Fast thermal response and high peak temperature can lead to unacceptable consequences such as media degradation and solution boiling.

For computational modeling purposes, a conservative approach was taken by assuming that the primary cooling mechanisms inside and outside of the column were conduction and natural convection, respectively, and that axial heat removal from the column was negligible relative to radial heat transfer. A two-dimensional transient heat conduction model was developed to assess the thermal performance of the packed column with loss of flow using the prototypic geometry as shown in Fig. 1 [5].

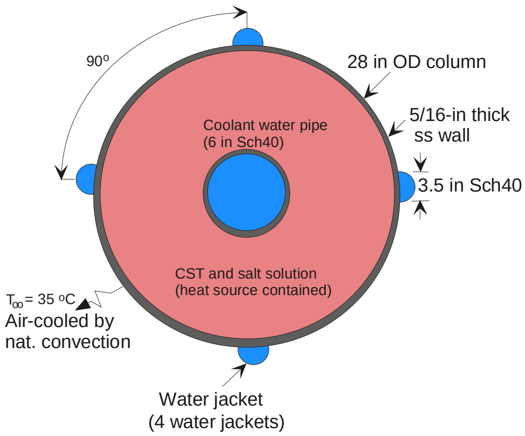

Figure 1. Modeling geometry and basic conditions used for the analysis.

The present work considers three basic cases. The first two cases are assumed to have no flow through the bed and involve internal heat transfer by conduction only for columns filled with salt solution. Heat transfer at the walls involves natural convection from the external wall boundary to the ambient air, with the assumption that the air serves as an infinite heat sink at constant temperature. These two cases are naturally-cooled stagnant columns with passive (Case 1) and active (Case 2) cooling systems, respectively. The last case, Case 3, considers the minimum expected salt solution flow (5 gpm) through the bed during normal operations for a bed containing the same heat source. In this case, it is assumed there is no heat loss at the column wall boundary and all heat transfer is through the mobile liquid phase with forced convection.

RESULTS

Transient two-dimensional heat conduction calculations have been performed to assess the thermal properties of cesium-saturated CST- and RF-packed ion exchange columns immersed in 6 M Na^+ salt solution under the baseline SCIX design. For Case 1, steady-state and transient temperature profiles within the cylindrical columns were determined for natural convection cooling under no process flow situations. This is considered a bounding conservative case. For Case 2 steady-state calculations were conducted to determine the maximum column temperatures under no process flow conditions but with active cooling (mixed natural/forced convection cooling). Case 3 quantified the steady state temperature responses under nominal operating conditions with 5 gpm liquid flow.

From the present modeling results, the main results are summarized:

- Under no process flow conditions with an inactive cooling system (Case 1) and columns suspended in unventilated ambient air, CST columns reach boiling temperatures within approximately 5 and 6 days for 55 and 35°C air temperatures, respectively. With 35°C ambient air under Case 1 conditions, the maximum temperature of the CST column reaches 60°C within ~1 day and 80°C within ~2 days. The temperature limit for CST stability is believed to be in the temperature range 50-80°C.
- Cesium-saturated RF-salt solution columns do not reach boiling temperatures under any conditions. The maximum temperatures of RF columns under Case 1 and 2 conditions are 87.6°C and 40.1°C, respectively. The temperature limit for RF stability is believed to be 65°C.
- Under no process flow conditions with a fully active cooling system (Case 2) and columns suspended in unventilated ambient air at 35°C, CST columns reach a maximum temperature of 63°C. Maximum steady-state temperatures predicted for CST columns with partial cooling involving only the central cooling pipe and only the water jackets are 80 and 114°C, respectively.
- The 6-in water cooling pipe located at the center of the column provides the most effective cooling mechanism under stagnant column conditions (cooling capacity ~33% of total heat load).
- Salt solution flow through the columns (Case 3) provides the most effective overall heat transfer mechanism. Maximum temperature differentials within the columns were less than 5°C in all cases with a liquid phase flow rate of 5 gpm.

REFERENCES

1. P. A. Taylor, C. H. Mattus, "Thermal and Chemical Stability of Baseline and Improved Crystalline Silicotitanate", ORNL/TM-2001/165 (December 2001).
2. C. E. Duffey, D. D. Walker, "Radiolytic, Thermal, and Physical Degradation Testing of Spherical Resorcinol-Formaldehyde Resin (U)" WSRC-TR-2005-00075 (July, 2006).
3. L. L. Hamm. S. E. Aleman, B. J. Hardy, W. D. King, and C. E. Duffey, "Ion Exchange Modeling for Removal of Cesium from Hanford Waste Using SuperLigR 644 Resin," Washington Savannah River Company, WSRC-TR-2003-00555 (February 2004).
4. F. G. Smith, III, "Modeling of Ion-Exchange for Cesium Removal from Dissolved Saltcake in SRS Tanks 1-3, 37 and 41", Washington Savannah River Company, WSRC-STI-2007-00315 (2007).

5. S. Y. Lee, "Heat Transfer Analysis for Fixed CST and RF Columns," Savannah River National Laboratory document, WSRC-STI-2007-00345, Rev. 0 (2007).